Use of $^{234}U/^{238}U$ Ratios to Measure *In Situ* Weathering Rates in the Hanford Vadose Zone

Katharine Maher, Donald J. DePaolo, and John N. Christensen
Contact: Katharine Maher, 510/642-9524, katem@eps.berkeley.edu

RESEARCH OBJECTIVES

Weathering rates of subsurface soil and rocks are difficult to quantify because of the difficulties in assessing the amount of reactive surface area and the thermodynamic potentials driving the chemical reactions. Mineral dissolution rates measured in the laboratory typically predict rates that are 2 to 4 orders of magnitude faster than estimates based on field measurements (White et al., 1996). Few field measurements are available, and most of those are on silicate soils. Almost no field measurements exist for deep vadose zone materials or for rocks in the saturated zone.

The uranium-series (U-series) isotope system can be used to measure reaction rates in aquifers and thick vadose zone environments. This approach is based on α-recoil of ^{234}Th atoms across grain boundaries, which enriches the pore fluid in ^{234}U. Dissolution of the solid phase releases mainly ^{238}U to the pore fluid, so that the $^{234}U/^{238}U$ ratio of the pore fluid is a measure of the local ratio of the dissolution uranium flux to the α-recoil flux (Tricca et al., 2001; DePaolo et al., 2003 submitted). The *in situ* reaction rate can be calculated from measurements of the $^{234}U/^{238}U$ isotopic ratio of interstitial fluids and solid phases, if the α- recoil flux can be estimated independently.

APPROACH

Uranium isotopes were measured for bulk sediment, sediment size fractions, pore fluids, the exchangeable fraction, and selected mineral phases of a 70 m vadose zone core at the Hanford Site, Washington, to estimate the mineral weathering rates and understand uranium distributions in the vadose zone. These measurements were performed using a Micromass IsoProbe multicollector ICP-MS at the Center for Isotope Geochemistry. A reactive transport model, aimed at comparing the U-series kinetics to those predicted by a multicomponent thermodynamic model, is being constructed to further evaluate the results.

ACCOMPLISHMENTS

The measured $^{234}U/^{238}U$ ratios for the vadose zone core yield weathering rates that are in general agreement with estimates based on other methods: approximately $10^{-6.4}$ yr^{-1} (Figure 1). These are the first measurements of uranium isotope compositions of the vadose zone pore waters and solid phases, and the first attempt to use these measurements to estimate weathering rates in the vadose zone.

SIGNIFICANCE OF FINDINGS

This study is the first to quantify reaction kinetics in the vadose zone. Perhaps the most significant contribution of this work is the development of an *in situ* method for determining precipitation/dissolution rates that, when coupled with other geochemical data, may help to decipher the mechanisms that control weathering in natural systems.

RELATED PUBLICATIONS

DePaolo et al., Rate of diagenetic reactions in deep-sea sediment: *In situ* measurement using $^{234}U/^{238}U$ of pore fluids. Geochim. Cosmochim. Acta., 2003 (submitted).

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Tricca, A., G.J. Wasserburg, D. Porcelli, and M. Baskaran, The transport of U- and Th- series nuclides in a sandy unconfined aquifer. Geochim. Cosmochim. Acta, 65, 1187–1210, 2001.

White, A.F., A.E. Blum, M.S. Schulz, T.D. Bullen, J.W. Harden, and M.L. Peterson, Chemical weathering of a soil chronosequence on granitic alluvium 1. Reaction rates based on changes in soil mineralogy. Geochim. Cosmochim. Acta, 60, 2533–2550, 1996.

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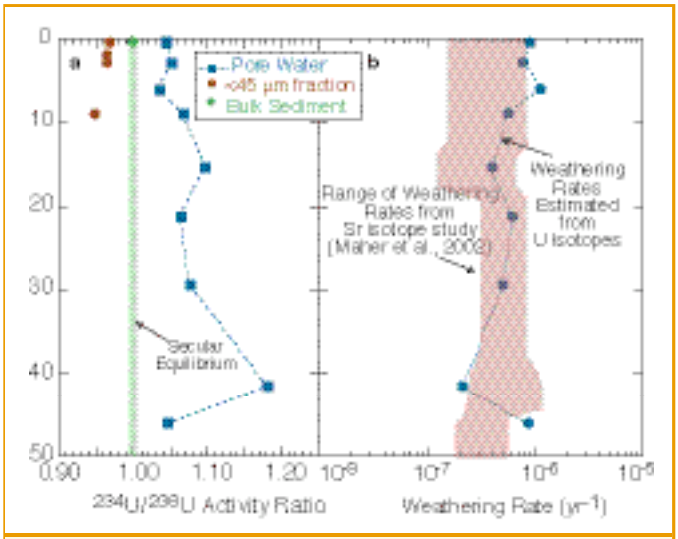

Figure 1. (a) U isotope data from the 299-W22-48 core, Hanford, Washington. (b) Comparison of estimated weathering rates derived from Sr and U isotope studies.